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(54) Title: ELECTRICALLY INSULATING FILM BACKING			
(57) Abstract			
<p>A halogen-free, electrically insulating film comprising a resin component containing: a) from 0 to 40 parts of a rubber selected from EP or EPDM rubber; b) correspondingly, from 60 to 100 parts of an ethylene vinyl acetate polymer; c) from 40 parts to 150 parts ethylene diamine phosphate per 100 parts of said resin component; and d) from 0.5 to 5 parts of an amino-functional silane coupling agent per 100 parts resin component; wherein a nonoriented film self-extinguishes in less than 5 seconds, has an elongation at break of at least 200 %, a dielectric strength of at least 1200 V/Mil, and said film has a stress-strain relationship such that a curve showing a first derivative of stress versus strain is positive over the entire curve, and a curve showing a second derivative of stress versus strain is negative over more than 50 % of said curve.</p>			

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ELECTRICALLY INSULATING FILM BACKINGBackground of the InventionField of the Invention

The invention relates to electrically insulative
5 polymeric films useful as conductive insulators and
electrical tape backings that contain no halogen
material.

Description of the Related Art

10 Various electrically insulative resins are known
in the art. Polyolefins have been used in various
resins, with and without flame retardancy in the
electrical industry.

Most insulative films which are used commercially,
15 and have both good flame retardancy and good physical
properties contain some vinyl chloride. Because of the
toxins produced when such compositions are burned,
either accidentally or when discarded, it is desirable
to reduce the halogen content as much as is possible,
20 particularly chlorine content. However, it is
difficult to attain both the flame retardancy and the
physical properties such as tensile and elongation in
an alternative product. This is especially true of
25 films being used for tape backings in the electrical
industry where the tapes must stretch and drape in a
certain manner to be acceptable for use.

U.S. Patent 5,017,637 discloses fire-retardant
thermoplastic compounds that are chemically
crosslinked, comprising 5-60% olefinic copolymers,
30 1-15% organopolysiloxane, and 20-85% flame retardant.
Preferred embodiments include a copolymer, and an
additional elastomer or ethylene copolymer. The
preferred elastomers are EP or EPDM copolymers.
Ethylene vinylacetate is also disclosed. Metal oxide
35 hydrates are used as the fire-retardant compound.

U.S. Patent 4,772,642 discloses a resin containing
polyolefins, preferably polypropylene. Ammonium
polyphosphate particles are used for flame retardance,
and a melamine resin encapsulates the particles.

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EP Patent 274,888 discloses a flame retardant halogen-free elastomer composition containing greater than 50% inorganic filler. Addition of a coupling agent is not disclosed.

- 5 EP Patent 391,336 discloses the use of a silane coupling agent and/or an olefinic synthetic rubber in a flame retardant composition of polypropylene and ammonium polyphosphate or melamine modified ammonium polyphosphate with one or more nitrogen-containing
10 organic compounds. The organofunctional group of the silane can be vinyl, chloro, amino or mercapto. Vinyl trimethoxysilane, vinyl triethoxysilane and 3-mercaptopropyl trimethoxy silane are preferred. Compositions with an olefinic synthetic rubber and
15 without silane are also disclosed.

JP Patent 04,139,241 discloses an olefinic resin blended with ammonium polyphosphate, a silane coupling agent, and olefinic synthetic rubber and a petroleum resin. An example of the silane is vinyl
20 trimethoxysilane. Advantages of this compound are high electrical resistance and high bleed resistance.

US Patent 5,130,357 discloses a flame retardant composition containing polypropylene as the major constituent, a silane coupling agent, and/or olefinic
25 synthetic rubbers, ammonium polyphosphate (APP) or melamine-modified APP and one or more nitrogen compounds, and optionally polyethylene resin, crosslinking agent and thiophosphate.

U.S. Patents 4,808,474 and 4,769,283 disclose a
30 pressure-sensitive adhesive tape backing having improved toughness comprising blends of crystalline isotactic polypropylene and compatible flexible polymers (in the former) or compatible ethylene containing polymers (in the latter), such polymers
35 including EPDM and/or EVA.

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U.S. 4,985,024 discloses a biodegradable pressure-sensitive adhesive tape backing comprising such a blend combined with an unsaturated elastomer.

U.S. Patent 5,134,012 discloses a fixing tape for 5 a disposable diaper comprising a plastics film layer, a polymer blend layer, and an adhesive layer. The plastics film layer may contain an EVA copolymer, and EP copolymer, or a combination thereof; the polymer blend layer contains two or more resins selected from 10 EVA, EP, and polyethylene.

U.S. Patent 3,941,859 discloses EPDM polymers physically blended with polyethylene and ethylene vinyl acetate copolymers having improved tensile strengths. Use as wire and cable insulation is disclosed.

15 However, these attempts to produce a halogen free film for the electrical industry, and especially for tape backings, have not been able to produce a halogen-free film with the required flame retardance and physical properties.

20 The present inventors have found that films comprising ethylene vinyl acetate (EVA) copolymers and an elastomer selected from ethylene propylene (EP) and ethylene propylene diene monomer (EPDM) rubbers and an effective amount of flame retardant phosphorous-
25 nitrogen containing intumescence fillers and an amino-functional silane coupling agent has tensile stress-strain behavior such that the first derivative of stress with respect to strain is positive for the entire curve, and a curve showing a second derivative
30 of stress-strain is negative over more than 50% of the curve.

Further, such films will provide electrical tape backings having the feel, and handling properties of the most popular poly(vinyl chloride) backings with no 35 halogen, which eliminates the release of toxic gases into the air, and also reduces negative environmental aspects from processing and disposal.

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Summary of the Invention

The invention provides a halogen-free, electrically insulating film comprising a resin component containing:

- 5 a) from 0 to 40 parts of a rubber selected from EP or EPDM rubber,
- b) correspondingly, from 60 to 100 parts of an ethylene vinyl acetate polymer,
- c) from about 40 parts to 150 parts of ethylene diamine phosphate per 100 parts of said resin component, and
- d) from 0.5 part to 5 parts of an amino-functional silane coupling agent per 100 parts of said resin component,
- 15 wherein a nonoriented film self-extinguishes in less than 5 seconds, has an elongation at break of at least 200%, a dielectric strength of at least 1200 V/Mil, and stress-strain behavior such that a curve showing a first derivative of stress-strain is positive over the entire curve, and a curve showing a second derivative of stress-strain is negative over more than 50% of said curve.
- 20 25

Insulating films of the invention are suitable for use as an electrical tape backing. Preferred

- electrical tape backings are halogen-free electrical tape backings comprising a resin component containing:
 - a) from 10 to 40 parts of a rubber selected from EP or EPDM rubber,
 - b) correspondingly, from 60 to 90 parts of an ethylene vinyl acetate polymer,
 - c) from 40 parts to 150 parts of ethylene diamine phosphate per 100 parts of said resin component, and
 - d) from 0.5 part to 5 parts of an amino-functional silane coupling agent, per 100 parts of said resin component,
- 30
- 35

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wherein a nonoriented film self-extinguishes in less than 5 seconds, has an elongation at break of at least 200%, a dielectric strength of at least 1200 V/Mil, and the stress-strain behavior described above.

5 The invention also provides electrical tapes comprising a halogen-free backing film, comprising a resin component containing:

- a) from 0 to 40 parts of a rubber selected from EP or EPDM rubber,
- 10 b) correspondingly, from 60 to 100 parts of an ethylene vinyl acetate polymer,
- c) from 40 parts to 150 parts of ethylene diamine phosphate per 100 parts of said resin component, and
- 15 d) from 0.5 part to 5 parts of an amino-functional silane coupling agent, per 100 parts of said resin component,

wherein said tape self-extinguishes in less than 5 seconds, has an elongation at break of at least about 20 200%, a dielectric strength of at least 1200 V/Mil, and a stress-strain curve as described above, and an adhesive coated on one major surface of said backing.

All weights, percents, parts, and ratios herein are by weight unless specifically noted otherwise.

25

Description of the Drawings

FIG. 1 shows the stress-strain curve for Example 1 and Comparative Example 1 films having thicknesses of 150 μm to 200 μm (6-8 mils).

30 As can be seen from the figure, there is dramatic improvement in mechanical properties for films containing the amino-functional silane. The yield point completely disappears, and the lower elongation of Example 1 is evidence of the improved adhesion 35 between the filler and the matrix polymers.

FIG. 2 shows the stress strain curves for films having various amounts of amino-functional silane.

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FIG. 3 shows stress-strain curves for films wherein the type of silane is varied. Films containing the amino functional silanes exhibit a dramatically changed curve as compared to vinyl functional silanes, 5 benzyl mercapto functional silanes, and methacryl functional silanes, demonstrating the reinforcing effect of the amino functional silanes.

FIG. 4 shows the stress-strain curves for electron beam irradiated films. Flame retardant properties are 10 not noticeably effected by the addition of differing types of silane coupling agents.

FIG. 5 shows the stress-strain curves of non-silane coupling agents which contain an amino functional group, i.e., amino-functional titanates and 15 zirconates.

FIG. 6 shows the stress strain curves of the effects of no silane, and two different levels of amino silane with various flame retardant fillers.

FIG. 7 shows the stress-strain curve of 20 compositions having non-phosphorous-nitrogen (P-N) type flame retardants.

FIG. 8. shows the stress-strain curves of various matrix polymers with P-N type flame retardants of this invention.

25 FIG. 9 shows the stress-strain curves at low temperatures of Example 1 of the invention and Comparative Example 3.

Detailed Description of the Invention

30 Compositions of the invention comprise a resin component containing at least one ethylene-vinyl acetate copolymer (EVA). Ethylene vinyl acetate copolymers useful in the films of the invention contain at least 10% by weight vinyl acetate, preferably at 35 least 15% by weight. The resin component may contain only ethylene vinyl acetate, or it may also contain a rubber selected from EP and EPDM rubbers; when such a

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rubber is present, the resin component may contain as little as 60 percent EVA copolymer.

Useful rubber polymers comprise from about 50% to 90% of ethylene, from 10% to 50% propylene, and from 0 5 to 3% diene.

Examples of suitable diene monomers include, but are not limited to, conjugated dienes such as isoprene, butadiene, 2,3-hexadiene, and the like, and nonconjugated dienes such as 1,4-pentadiene, 1,5-
10 hexadiene, 2, 5-dimethyl-1, 5 hexadiene, 1, 4-hexadiene and the like; cyclic dienes such as cyclopentadiene, cyclohexadiene, dicyclopentadiene, 3-methyl-tricyclo(5,2,1)-3,8-decadiene, and the like, and alkenyl norbornenes such as 5-ethylidene-2-norbornene, 2-
15 methallyl-5-norbornene, and the like. These polymers are easily prepared by well known solution or suspension polymerization techniques.

Insulating films of the composition comprise an effective amount of ethylene diamine phosphate as the
20 flame-retardant agent, such as that available commercially from Albright & Wilson.

Insulating films of the invention also comprise an amino silane coupling agent. Useful amino silanes include, but are not limited to, N-beta-(amino-ethyl)
25 gamma-aminopropyl trimethoxy silane and aminopropyl triethoxy silane and trimethoxy silane. Films of the invention contain from 0.05 part to 5 parts amino-functional silane coupling agent, preferably from 0.1 part to 2 parts per 100 parts resin component.

30 Films of the invention may also comprise conventional additives such as reinforcing fillers, pigments such as carbon black, and TiO₂, dyes, ultraviolet stabilizers, plasticizers, fungicides, extenders, waxes, antioxidants, and the like, in
35 amounts known to those skilled in the art.

Other useful fillers include fumed silica, calcium and magnesium carbonates, calcium and barium sulfates,

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aluminum silicates, and the like, which may be included in small amounts, such that they do not interfere with the physical properties required.

- Films of the invention are useful as insulative
- 5 wire and pipe coatings, as insulative backings for multilayer films, and especially, as electrical tape backings. Such films would also be useful for various molded and extruded items such as shoe soles, shower curtains, kitchen ware and the like.
- 10 The films of the invention are made by physically mixing the rubber, the ethylene vinyl acetate copolymer, and the ethylene diamine phosphate, along with any additives in a mill, mixer or extruder. The mixing conditions are not critical, and such processes
- 15 are well known to one skilled in the art.

Films of the invention have a stress-stain behavior such that a curve of the first derivative stress-strain relationship has a wholly positive slope. This is surprising, as most olefin-based films have at least some portion of the slope which is negative.

20 Further, a curve of the second derivative shows that it is mostly negative. That means that these films do not have an inflection point below 200% elongation.

Electrical tapes backed with films with these

25 characteristics demonstrate a vastly improved handling characteristic over the prior art halogen-free films; i.e., they approximate the stretch and "drape" characteristics of vinyl chloride backed electrical tapes. This is extremely critical for proper

30 insulation and sealing of repaired cables and connections, as well as for acceptance by persons skilled in electrical maintenance and repair.

Tapes comprising backings of the invention have an improved low temperature performance as compared to

35 prior art poly(vinyl chloride) backings. PVC film becomes very stiff, exhibits a yield point and is brittle at the low temperature. The filled blend

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retains good elongation and mechanical properties even at temperatures of -20°C. Further, vinyl does not have a stress-strain curve with a wholly positive slope at such low temperatures, whereas films of the invention 5 do have a stress-strain curve which retains a positive slope even at -20°C.

Electrical tapes of the invention have at least one side of the film backing coated with an adhesive. The adhesive may be any conventional adhesive known in 10 the art, including but not limited to, natural rubber, thermoplastic elastomers, such as block copolymers, thermoset adhesives, acrylic adhesives, silicone adhesives and the like. The adhesive may further comprise such conventional additives as tackifiers, 15 plasticizers, pigments, fillers, initiators, crosslinking agents, and the like, as desired.

The following examples are meant to be illustrative and should not be construed as limiting the scope of the invention, which is defined only by 20 the claims. One skilled in the art would be able to create variations which would be within the spirit of the invention. Comparative examples are indicated by the use of the letter "C" in front of the example number.

25

Test Methods

Flame Retardance

Flame retardance was tested by the ASTM D1000 test method. This test involves wrapping a film strip 30 around a wire with a 50% overlap and repeating with another film strip in the opposite direction. The wrapped wire is exposed to an open flame for 30 seconds. The flame is removed and the burn time of the film is measured. Desirable flame retardance would be 35 exhibited by a material that does not begin to burn, or self extinguishes in less than 5 seconds.

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Tensile Test

Tensile strength was tested by the ASTM D1000 test method.

5		<u>Glossary of Materials</u>
	Elvax™ 470	poly(ethylene vinyl acetate) copolymer; 18% vinyl acetate; available from DuPont
	Epsyn™ 7506	ethylene-propylene terpolymer; 77% ethylene; 5.5C/100C unsaturated; available from Copolymer
10		low density polyethylene 1017; available from Chevron
	LDPE	amorphous polypropylene-polybutylene copolymer; available from Eastman
	PP-PB	experimental ethylene methyl acrylate-acrylic acid terpolymer; available from Exxon
15 LDX 314		poly(ethylene vinyl alcohol); 27% vinyl alcohol; available from EVALCO
	EVOH 27	ammonium polyphosphate based flame retardant filler; available from Hoechst-Celanese
20 IFR 10		ammonium polyphosphate flame retardant filler; available from Hoechst-Celanese
	IFR 23	ammonium polyphosphate based flame retardant filler; available from Hoechst-Celanese
25 Phoscheck P40		ammonium polyphosphate based flame retardant filler; available from Monsanto
	Exolit 422	ammonium polyphosphate based flame retardant filler; available from Hoechst-Celanese
30 EDAP		ethylene diamine phosphate; available from Albright & Wilson
	DE83R	decabromodiphenyl oxide flame retardant filler; available from Great Lakes Chem. Corp.
35 Solem 932		alumina trihydrate; available from J.M. Huber

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- EVA85H antimony trioxide concentrate in poly(ethylene vinyl acetate); available from Laurel
- A0750 aminopropyltriethoxy silane; available from Union Carbide
- 5 A151 vinyltriethoxy silane; available from Union Carbide
- A1100 aminopropyltrimethoxy silane; available from Union Carbide
- 10 A1120 N-beta-(aminoethyl)-gamma-aminopropyltrimethoxy silane; available from Union Carbide
- 1130 triamino-functional silane; available from Union Carbide
- 15 A174 gamma-methacryloxypropyltrimethoxy silane; available from Union Carbide
- M8500 3-mercaptopropyltrimethoxy silane; available from Huls Petrarch
- Z6032 N-[2(vinyl benzyl amino)-ethyl]-3-aminopropyltrimethoxy silane; available from Dow Corning
- 20 Lica 44 neopentyl(diallyl)oxy, tri(N-ethylenediamino)ethyl titanate; available from Kenrich
- 25 Lica 97 neopentyl(diallyl)oxy, tri(m-amino)phenyl titanate; available from Kenrich
- L44/H 2,2(bis-2-propenolatomethyl)butanlato, tri(N-ethylaminoethylamino) titanate; available from Kenrich
- 30 NZ44/H 2,2(bis-2-propenolatomethyl)butanlato, tri(N-ethylaminoethylamino) zirconate; available from Kenrich
- L97/H 2,2(bis-2-propenolatomethyl)butanlato, tri(m-amino) phenyl titanate; available from Kenrich
- 35 NZ97/H 2,2(bis-2-propenolatomethyl)butanlato, tri(m-amino) phenyl zirconate; Kenrich

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- Irganox 1010 hindered phenolic antioxidant; available from Ciba Geigy
 Irganox 1035 hindered phenolic antioxidant; available from Ciba Geigy

5

Examples

Examples of typical polymer blend compositions with and without the amino-functional silane coupling agent are provided by the formulations in Table 1.

- 10 Compositions were mixed in a Brabender™ rheometer using a small batch mixing head with high shear paddles at 105°C for 5 minutes until a uniform dispersion of the polymer and filler components was achieved. Blends were pressed between heated platens to form films.

15

Table 1

Materials	Example 1 (Parts)	Example C1
Exvax™ 470	80	80
Epsyn™ 7506	20	20
EDAP	50	50
A0750	0.15	-----
Irganox™ 1010	0.15	0.15

- The tensile properties of Example 1 and C1 films having a thickness of 150 µm to 200 µm (6-8 mils) of Examples 1 and C1 are depicted in Figure 1. As can be seen from the figure, there is dramatic improvement in mechanical properties for films containing the amino-functional silane. The yield point completely disappears, and the lower elongation of Example 1 is evidence of the improved adhesion between the filler and the matrix polymers.

The shape of the curve for the composition of Example 1 more closely resembles the behavior of

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plasticized poly(vinyl chloride) (PVC) which is highly desirable for films used in tapes for the electrical industry.

5

Examples C2 and 2-4

Blends were prepared in the same manner as described previously except containing varying amounts of amino-functional silane. Figure 2 demonstrates the effect of the amount of amino-functional silane on the 10 mechanical properties for the following compositions listed in Table 2.

Table 2

Material	Ex. 2	Ex. 3	Ex. 4	Ex. C2
Elvax™ 470	80	80	80	80
Epsyn™ 7506	20	20	20	20
EDAP	50	50	50	50
A1100	0.3	0.6	1.0	0
Irganox™ 1010	0.15	0.15	0.15	0.15

20

As addition of amino-functional silane increases, the shape of the stress-strain curve remains approximately the same although slight increases in tensile strength and reductions in ultimate elongation 25 occur with higher silane contents. The stress-strain behavior of plasticized vinyl (PVC) film is shown for comparison of the relative shapes of the curves (Example C3).

Compositions for several blends containing a 30 phosphorous-nitrogen flame retardant (EDAP) and various coupling agents including a composition with no coupling agent were hot melt mixed and pressed into films as described previously and are listed in Table 3.

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Examples 3-5 and C3-C5

These examples were made similar to Example 1, i.e., with 80 parts Elvax™ 470, 20 parts Epsyn™ 7506, 50 parts EDAP, and 0.15 part Irganox™ 1010. However, 5 the types of silane coupling agent were varied. The Example numbers and types of silane are listed below in Table 3.

Table 3

Silane	Ex 5	Ex 6	Ex 7	Ex C4	Ex C5	Ex C6
A1100	1					
A1120		1				
A1130			1			
A174				1		
M8500					1	
Z6032						1

Tensile stress-strain curves are shown in Figure 3 for films of the compositions listed in Table 3 and 20 film C1 shown in Table 1. The amino-functional silanes change the shape of the stress-strain curve dramatically compared to the vinyl, benzyl, mercapto and methacryl functional silanes. The Z6032 silane does contain an amino functional group, but it is 25 centrally located within the molecule and, for stearic considerations, is not freely accessible to interact with the polymer. The primary functionality of this silane is vinyl. These non-amino silanes, including the Z6032, may have some reinforcing effect over 30 compositions without silane, but not nearly the effect obtained with amino-functional silane coupling agents.

The dry and wet dielectric properties and the flame retardance of these compositions with the

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addition of various coupling agents and without coupling agent are listed in Table 4.

Table 4

5	<u>BURN TIME</u> <u>Seconds</u>	<u>DIELECTRIC STRENGTH</u> <u>(V/mil)</u>	
		<u>DRY</u>	<u>WET</u>
<u>EXAMPLE</u>			
	5	1653	537
	6	1534	518
10	7	1361	541
	C4	1756	535
	C5	1856	542
	C6	1589	536
	C1	1327	522

15

The dielectric properties of compositions using most types of coupling agents are improved over compositions without coupling agent. No advantage of amino-functional silanes, compared to other silanes, is 20 evident here. Flame retardant properties are not noticeably affected by the addition of different types of silane coupling agents. Amino-silane coupling agents perform similarly to other silanes in dielectric properties and flammability tests of these compounds.

25 Films of the compositions listed in Table 3 were exposed to electron beam radiation at a dose of 15 megarads. Tensile properties of irradiated samples are shown in Figure 4 and burn time and dielectric strength of irradiated samples are shown in Table 5.

30

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Table 5

<u>EXAMPLE</u>		<u>BURN TIME</u>	<u>DIELECTRIC STRENGTH</u>	
			<u>DRY</u>	<u>WET</u>
			<u>(V/mill)</u>	
5	5	1	1597	527
	6	2	1836	542
	7	2	1259	532
	C4	1	1940	523
	C5	1	1654	515
10	C6	2	1359	543
	C1	1	1079	525

Irradiation of these compositions changes the tensile properties by crosslinking the polymeric matrix, but the effect of the amino-functional silane on the tensile properties is still obvious compared to irradiated compositions containing no coupling agent. Although the curves are shifted together upon irradiation, close examination reveals that blends with non-amino functional silanes have an inflection point, a change from negative to positive second derivative of stress with respect to strain, below 200% elongation.

- Blends containing amino-functional silane exhibit an inflection point after irradiation, which is characteristic of crosslinked materials, but this occurs at elongations higher than 200%. Tensile properties of irradiated films containing non-amino functional silanes do not show the reinforcement seen with amino-functional silane coupling agents.
- Irradiated samples containing silane coupling agents demonstrate improvements in dry dielectric strength over samples without silane coupling agents, and amino silanes appear to perform similarly to other silanes, as was noted in non-irradiated samples.
- Irradiation has no obvious effect on flame retardance or dielectric properties.

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Coupling agents with amino-functionality, other than silane coupling agents, do not demonstrate the dramatic improvement in tensile stress-strain behavior. Blends containing amino-functional titanates and 5 zirconates are described in Table 6 and tensile properties of hot melt mixed and pressed films are shown in Figure 5.

Comparative Examples C7-C12

10 These Examples were made similar to Example C1, i.e., with 80 parts Elvax™ 470, 20 parts Epsyn™ 7506, 50 parts EDAP, and 0.15 part Irganox™ 1010. However, these examples use amino-functional titanates and zirconates in place of amino functional silanes of 15 compositions of the invention as coupling agents. The coupling agents are listed for each Example in Table 6.

Table 6

Ex. No. / Ingrid	Ex. C1	Ex. C7	Ex. C8	Ex. C9	Ex. C10	Ex. C11	Ex. C12
LICA 44	0	0.3					
LICA 97			0.3				
L44/H				0.3			
NZ44/H					0.3		
L97/H						0.3	
NZ97/H							0.3

Addition of these non-silane amino-functional coupling agents reduces the elongation of the 30 compositions and increases the tensile values somewhat, similar to non-amino silane coupling agents, but does not dramatically improve the tensile values or the shape of the stress-strain curve as does the amino functional silane coupling agents.

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Comparative Examples C13-C26 and Examples 8 and 9

Compositions containing various P-N flame retardant fillers are listed in Table 7. The effect of 0.3 phr and 1.0 phr of an amino-functional silane on 5 these compositions is also shown in Figure 6. EDAP is the only flame retardant that demonstrates the desired improvement in stress-strain properties. The other systems show very little change in tensile stress-strain properties with the addition of amino-functional 10 silane coupling agent.

Most of the commercial phosphorous-nitrogen type flame retardants (including those tested) are composed primarily of ammonium polyphosphate. EDAP is an exception. EDAP has tri-hydroxyl functionality that 15 imparts some level of acidity to this filler. Without wishing to be bound by theory, it is believed that this causes the EDAP to be more reactive than the APP compounds with the hydrolyzable groups of the silane. Infrared analysis was conducted on samples of EDAP, 20 EDAP in EVA/EPDM and EDAP in EVA/EPDM with amino-functional silane. The spectra indicate no chemical reaction between the EVA and the EDAP with or without the amino-functional silane. Hydrogen bonding would not be detectable as a chemical reaction.

25

Comparative Examples C27-C36

Films of flame retarded compositions that do not contain P-N type flame retardants were prepared that contain various types of silane coupling agents. These 30 are described in Table 8 and the tensile properties of these films are shown in Figure 7. The shape of the stress-strain curve is not affected by the different type of functionality of the silane coupling agent, although the addition of a silane of any type provides 35 a small improvement in tensile properties when compared with the properties of compositions containing no silane coupling agent.

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Table 7

Example No. Ingredients	Elvax™470	Epsyn™7506	EDAP	IFR 23	IFR 10	A1100	Irganox™
Ex. C1	80	20	50	-	-	0.15	
Ex. 8	80	20	50	0	0.3	0.15	
Ex. 9	80	20	50	-	1	0.15	
Ex. C13	80	20	-	50-	-	0.15	
Ex. C14	80	20	-	50	0.3	0.15	
Ex. C15	80	20	-	50	1	0.15	
Ex. C16	80	20	-	50	-	0.15	
Ex. C17	80	20	-	50	0.3	0.15	
Ex. C18	80	20	-	50	1	0.15	

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Table 7 Continued

Example No. Ingredient	Elvax™ 470	Epsyn™ 7506	P40	IFR 24	Exolit™ 422	A1100	Irganox™ 1010
Ex. C19	80	20	50	-		0	0.15
Ex. C20	80	20	50	-		0.3	0.15
Ex. C21	80	20	-	50		-	0.15
Ex. C22	80	20	-	50		0.3	0.15
Ex. C23	80	20	-	50		1	0.15
Ex. C24	80	20	-	-	50	-	0.15
Ex. C25	80	20	-	-	50	0.3	0.15
Ex. C26	80	20	-	-	50	1	0.15

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Table 8

Ex. No.	Elvax™	Epsyn™	SOLEM	DE83R	EVA85H	Irganox™	A1100	A151	A174	M8500
Ingrd.	470	7506	932			1010				
Ex. C27	80	20	60	-	-	0.15		-	-	
Ex. C28	80	20	60	-	-	0.15	0.3	-	-	
Ex. C29	80	20	60	-	-	0.15	-	0.3	-	
Ex. C30	80	20	60	-	-	0.15	-	-	0.3	
Ex. C31	80	20	60	-	-	0.15	-	-	-	0.3
Ex. C32	80	20	-	20	8	0.15	-	-	-	
Ex. C33	80	20	-	20	8	0.15	0.3	-	-	
Ex. C34	80	20	-	20	8	0.15	-	0.3	-	
Ex. C35	80	20	-	20	8	0.15	-	-	0.3	
Ex. C36	80	20	-	20	8	0.15	-	-	-	0.3

Comparative Examples C37-C40 and Examples 10-13

Compositions with a P-N type flame retardant and different matrix polymer materials were prepared and 5 pressed into films. Table 9 shows these compositions and the legend states whether the matrix polymer is capable of forming hydrogen bonds.

Figure 8 shows the tensile stress-strain curves for these blends. The polymeric materials that are 10 capable of forming hydrogen bonds demonstrate greatly improved mechanical properties with the addition of aminofunctional silane, i.e., elimination of a yield point and higher tensile values. The polymeric materials that are not capable of forming hydrogen 15 bonds do not show this type of improvement with addition of aminosilane. Reduced elongation is evident in all blends containing silane coupling agent. Without wishing to be limited by theory, it is believed that the amino functionality of the silane provides 20 dramatic improvements in mechanical properties for polymeric matrices capable of forming hydrogen bonds.

The PVC films used widely in electrical tapes display excellent ambient stress-strain properties which can be an indication of the films handling 25 behavior. Films of this invention have similar properties at room temperature, and also have superior low temperature stress-strain properties compared to PVC films.

Figure 9 shows ambient and low temperature stress-30 strain behavior for PVC film and low temperature stress-strain behavior for the material of Example 1. The shape of the Example 1 film shows no yield point and more closely resembles the ambient PVC behavior while the low temperature PVC curve has a yield point, 35 very low elongation, and very high ultimate tensile strength. At low temperatures, films of this invention have handling characteristics superior to PVC.

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Table 9

Ex. No.	Exavax™ Ingred.	Epsyn™ 7506	EDAP	A1100	Irganox™ 1010	LDX	Bynel™ 3048 ¹	PP-PB ^{2,3}	LLDPE ²	EVOH	Surlyn 2714	9020 ²
5	Ex. 10	80	20	50	0	0.15	100	-	-	-	-	-
Ex. 11	80	20	50	1	0.15	100	-	-	-	-	-	-
Ex. 12	80	20	50	0	0.15	-	100	-	-	-	-	-
Ex. 13	80	20	50	1	0.15	0	100	-	-	-	-	-
Ex. C37	80	20	50	0	0.15	-	-	100	-	-	-	-
Ex. C38	80	20	50	1	0.15	-	-	100	-	-	-	-
10	Ex. C39	80	20	50	0	0.15	-	-	-	100	-	-
Ex. C40	80	20	50	1	0.15	-	-	-	-	100	-	-
Ex. C41	80	20	50	0	0.15	-	-	-	-	-	100	-
Ex. C42	80	20	50	1	0.15	-	-	-	-	-	100	-
Ex. C43	80	20	50	0	0.15	-	-	-	-	-	-	100
15	Ex. C44	80	20	50	1	0.15	-	-	-	-	-	100

¹ Capable of forming H-bonds ² Not capable of forming H-bonds³ Not able to make films with this material; no mechanical integrity⁴ Not able to make films with this material; materials formed crosslinked thermoset

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What is Claimed is:

1. A halogen-free, electrically insulating film comprising a resin component containing:

5 a) from 0 to 40 parts of a rubber selected from EP or EPDM rubber,

b) correspondingly, from 60 to 100 parts of an ethylene vinyl acetate polymer, and

c) from 40 parts to 150 parts of ethylene diamine phosphate per 100 parts of said resin component, and

d) from 0.5 part to 5 parts of an amino-functional silane coupling agent per 100 parts of said resin component,

15 wherein a nonoriented film self-extinguishes in less than 5 seconds, has an elongation at break of at least 200%, a dielectric strength of at least 1200 V/Mil, and stress-strain behavior such that a curve showing a first derivative of stress-strain is positive over the 20 entire curve, and a curve showing a second derivative of stress-strain is negative over more than 50% of said curve.

2. A halogen-free electrical tape backing
25 comprising an insulating film according to claim 1.

3. An electrical tape comprising an adhesive and a backing film, said backing film being a halogen-free, insulating film according to claim 1.

30

4. A halogen-free electrically insulating film according to any of claims 1 through 3 wherein said ethylene diamine phosphate comprises from 40 parts to 80 parts per 100 parts of said resin component.

35

5. A halogen-free electrically insulating film according to any of claims 1 through 4 wherein said

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amino-functional silane coupling agent comprises from 0.1 part to 2 parts per 100 parts of said resin component.

- 5 6. A halogen-free electrically insulating film according to any of claims 1 through 5 wherein said amino-functional silane coupling agent is selected from the group consisting of N-beta-(amino-ethyl) gamma-aminopropyl trimethoxy silane and aminopropyl 10 triethoxy silane and trimethoxy silane.

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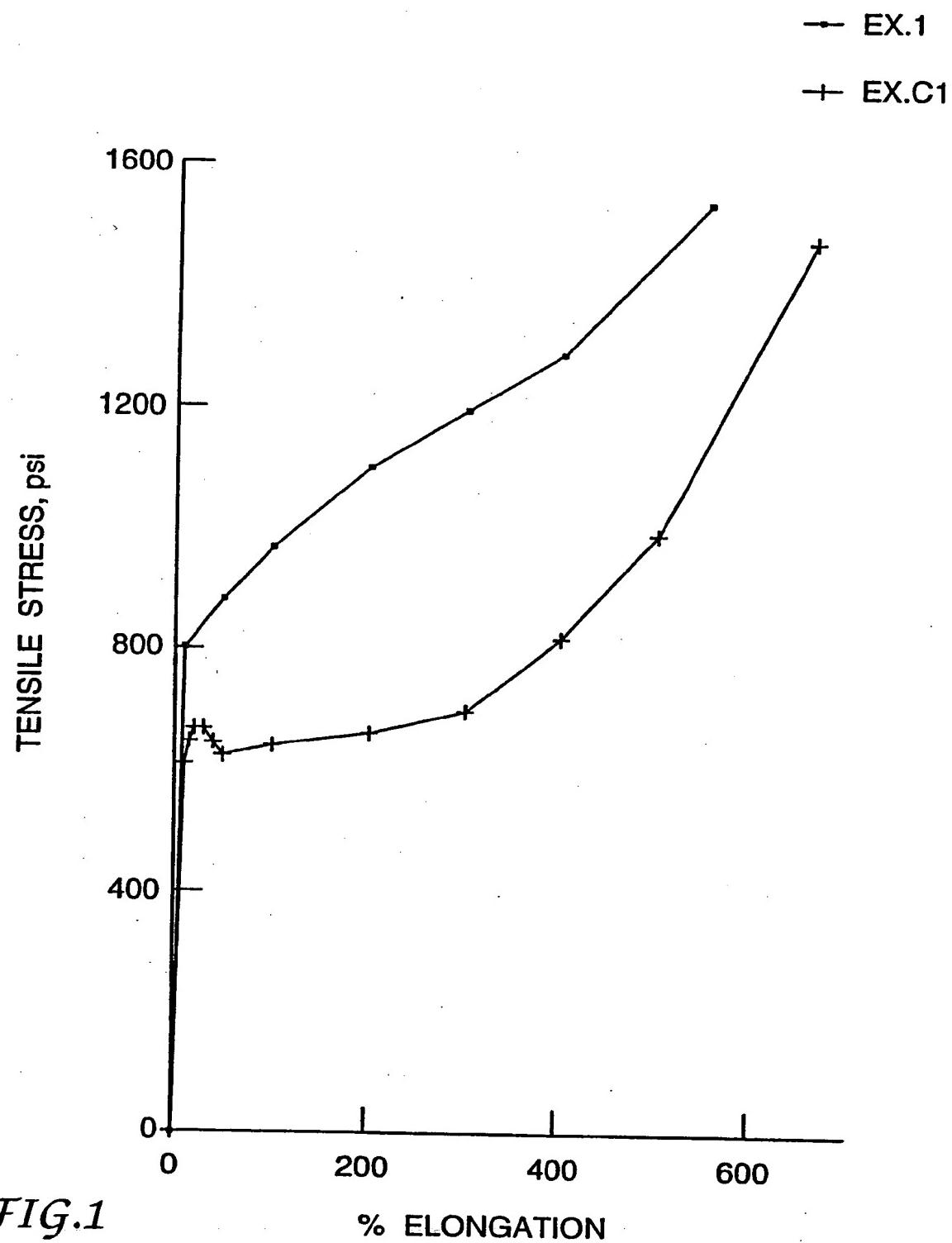


FIG.1

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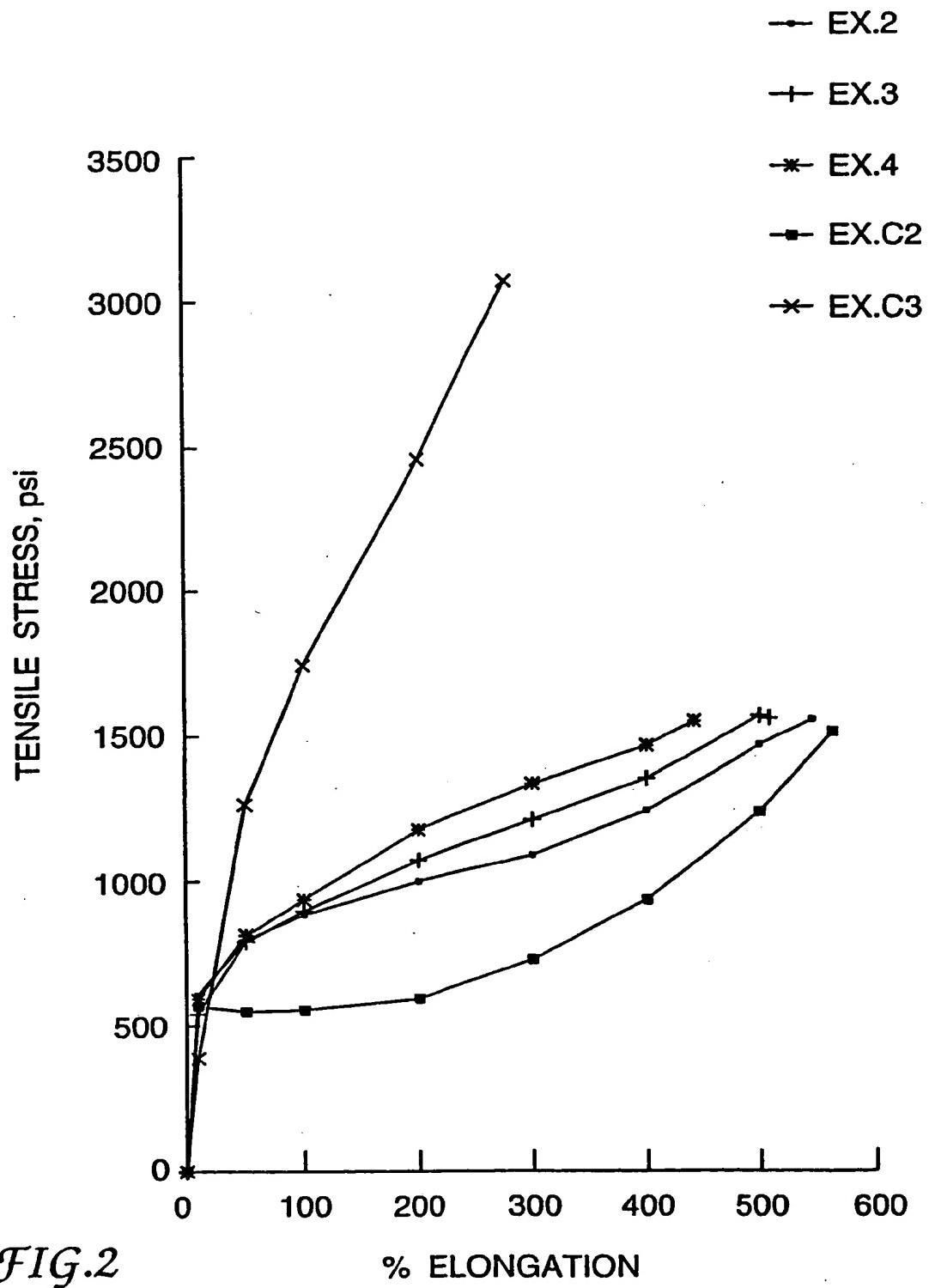


FIG.2

% ELONGATION

3/18

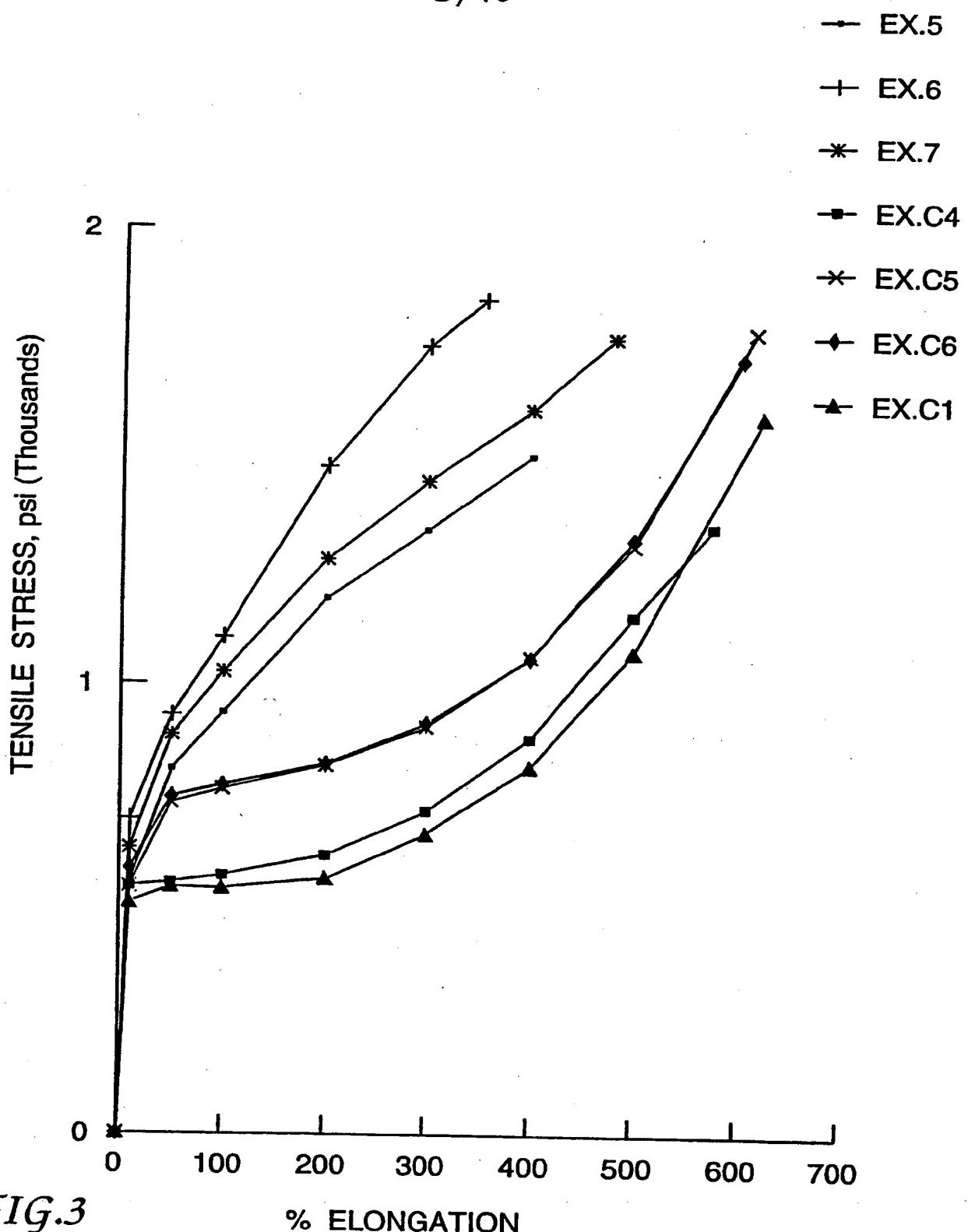


FIG.3

% ELONGATION

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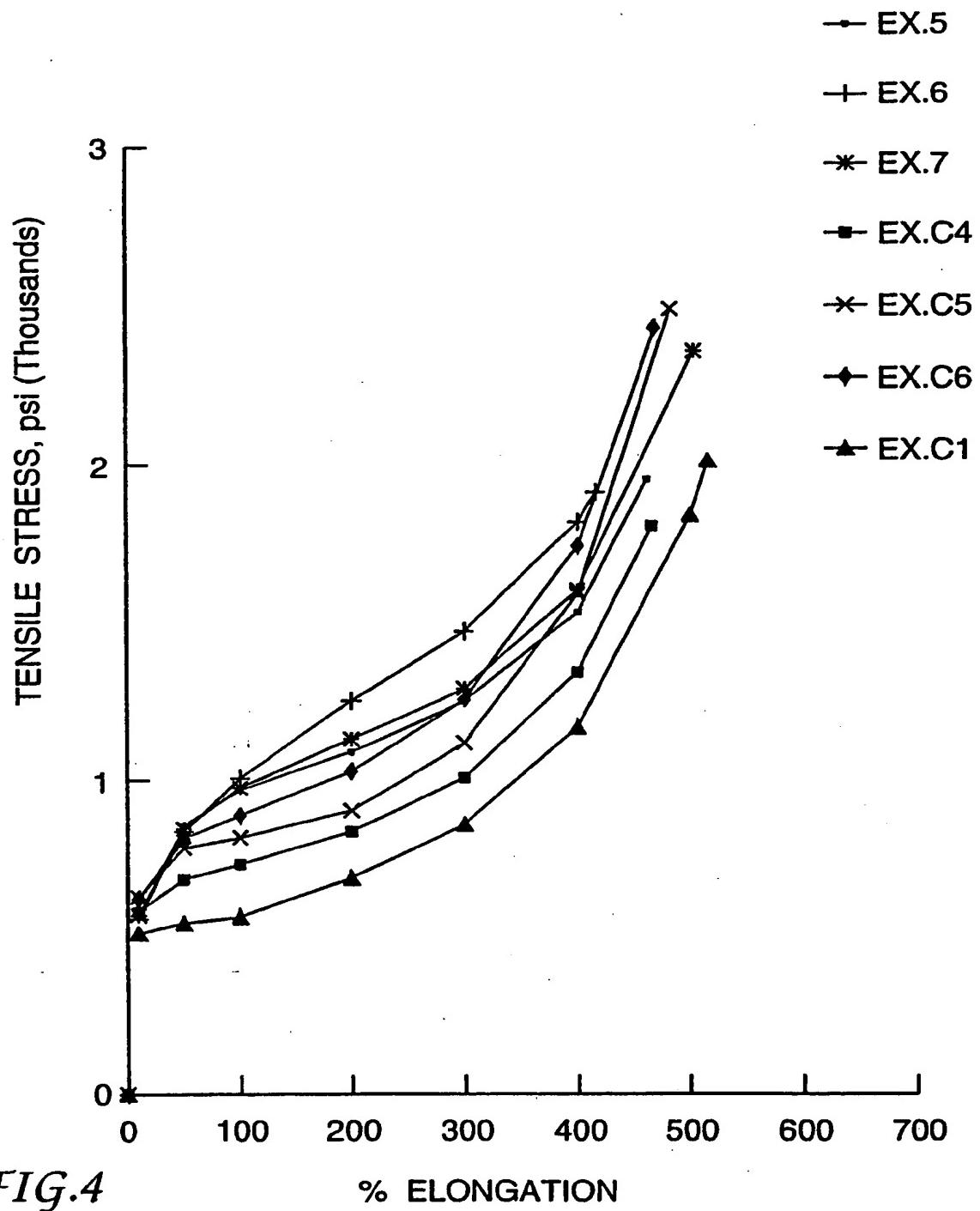


FIG.4

% ELONGATION

5/18

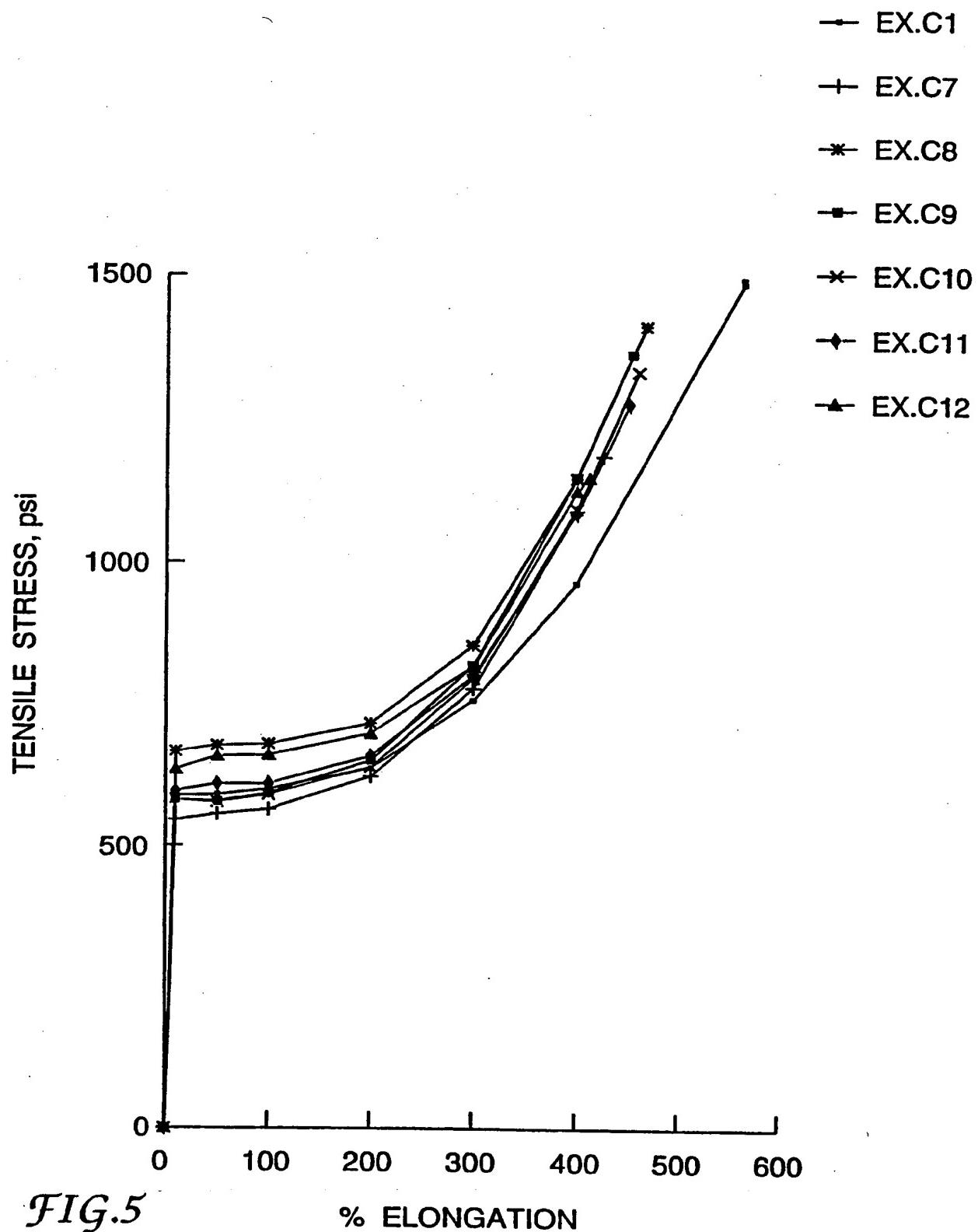


FIG.5

% ELONGATION

6/18

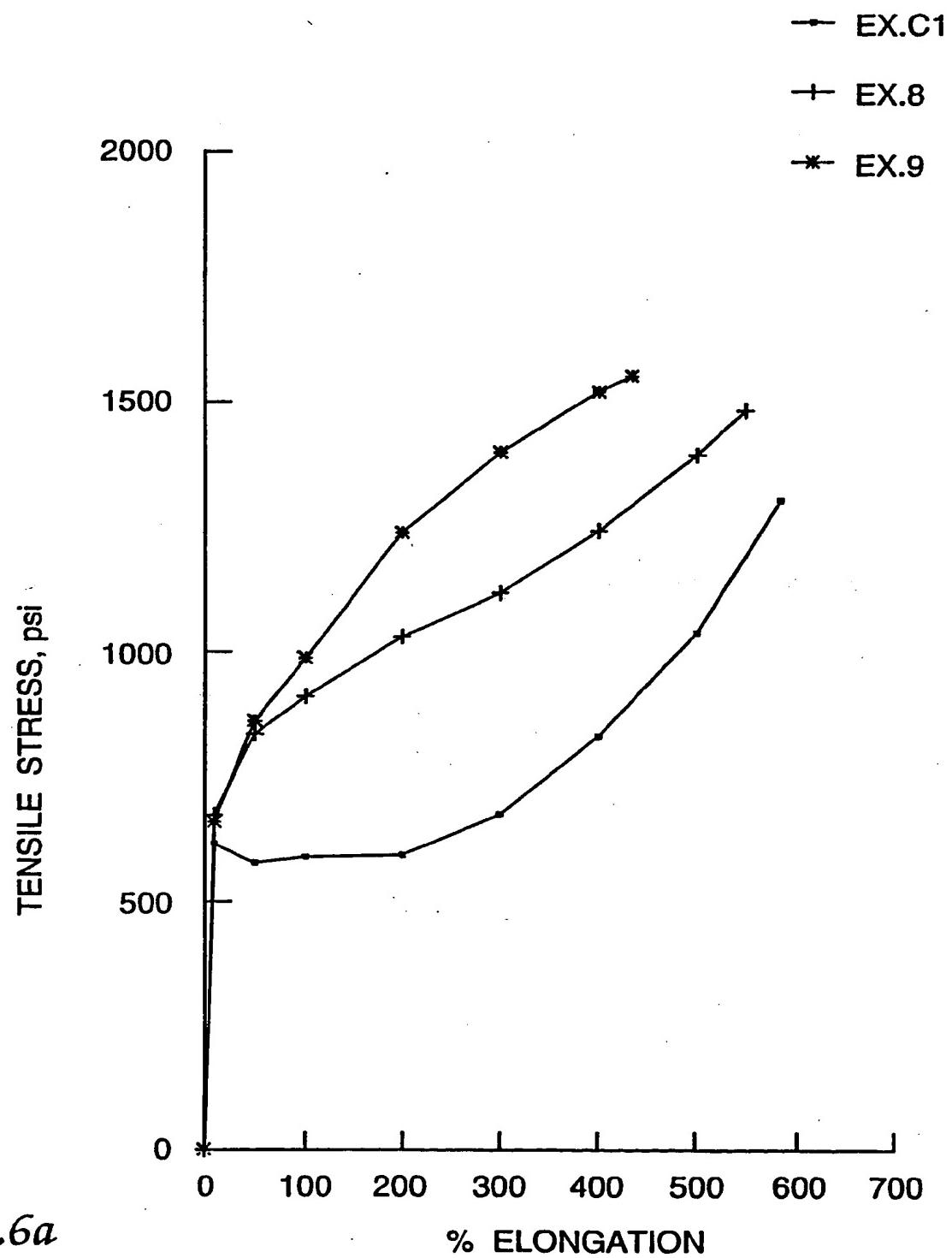


FIG.6a

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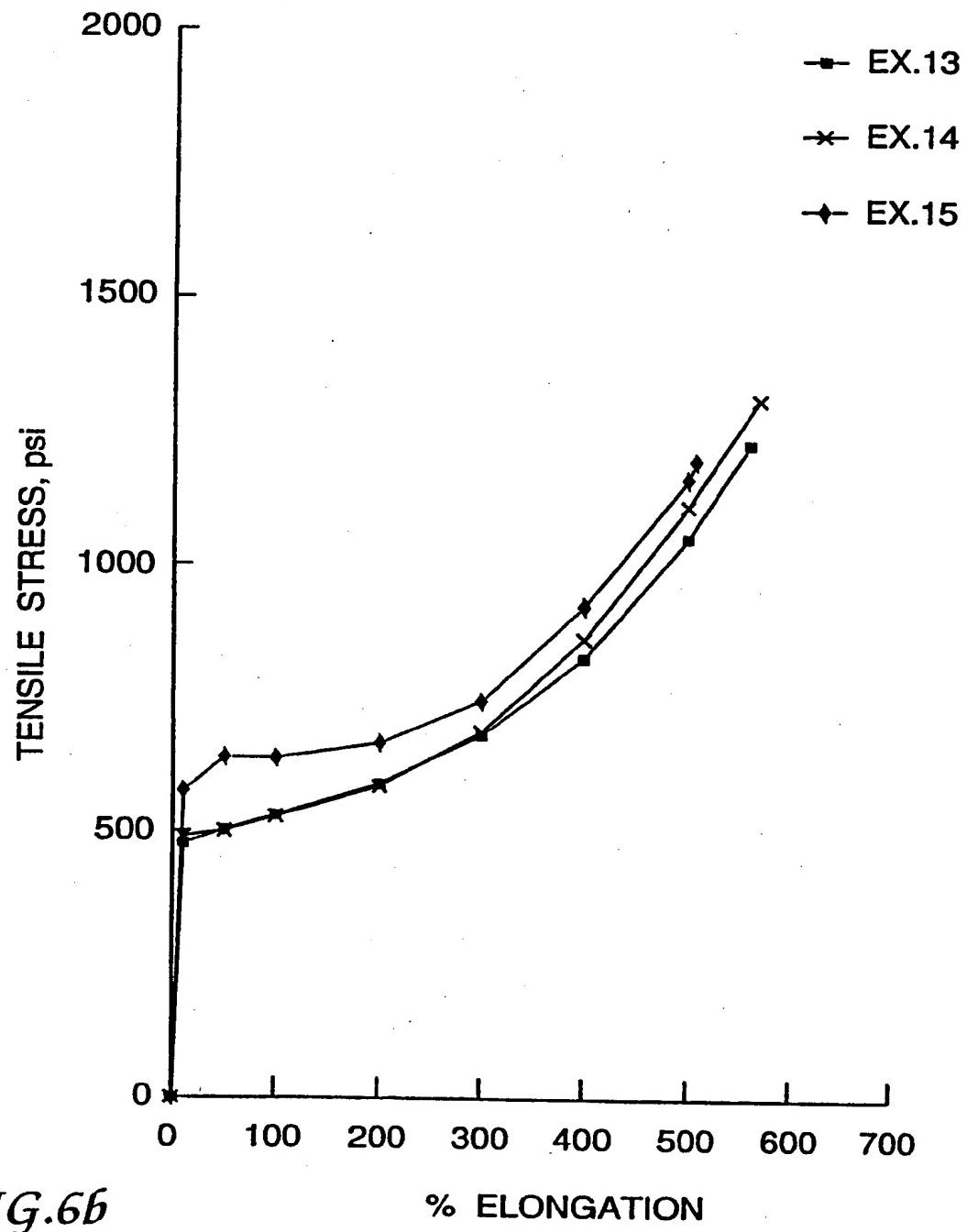


FIG.6b

% ELONGATION

8/18

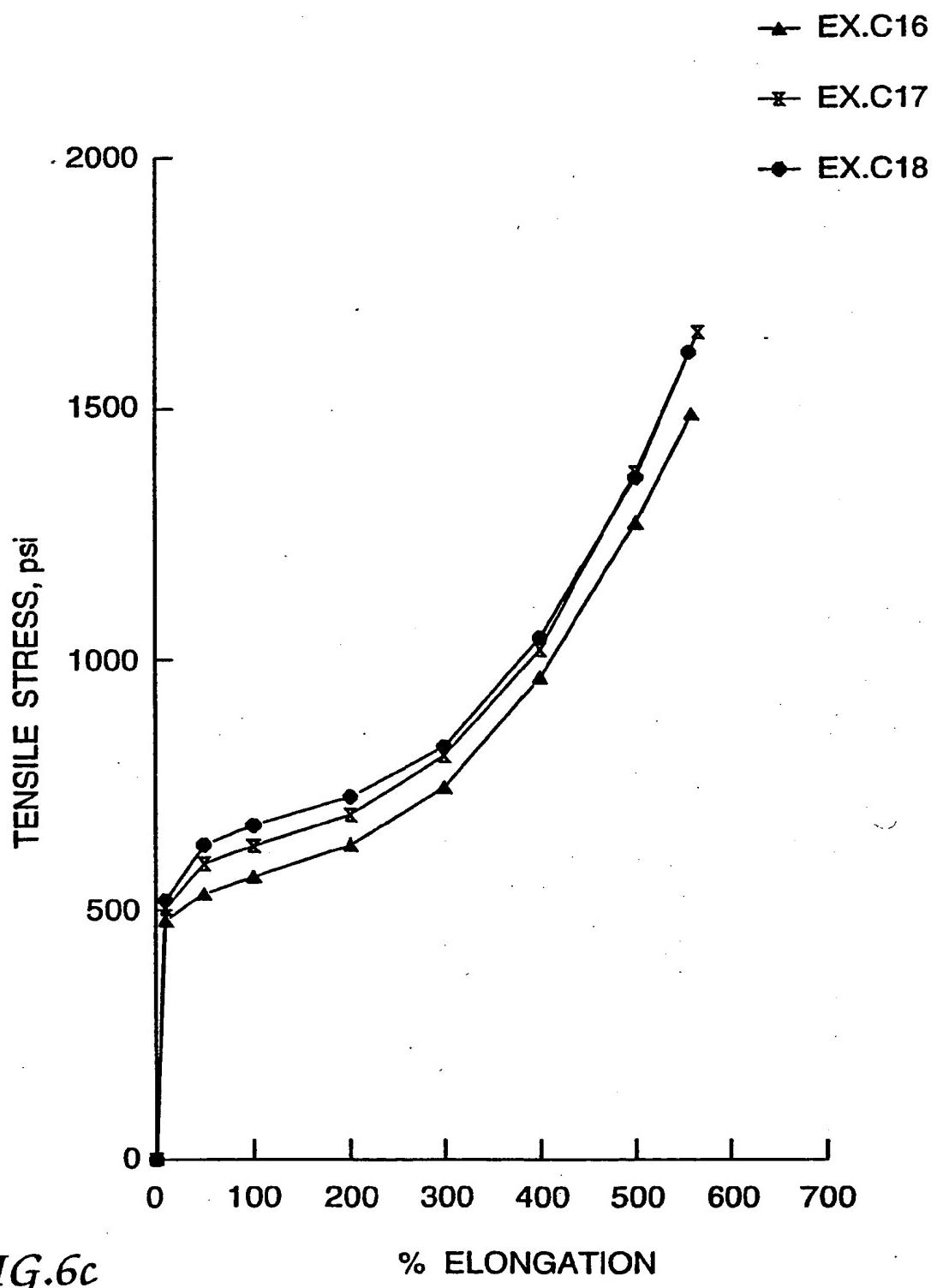


FIG.6c

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EX.C20
EX.C19

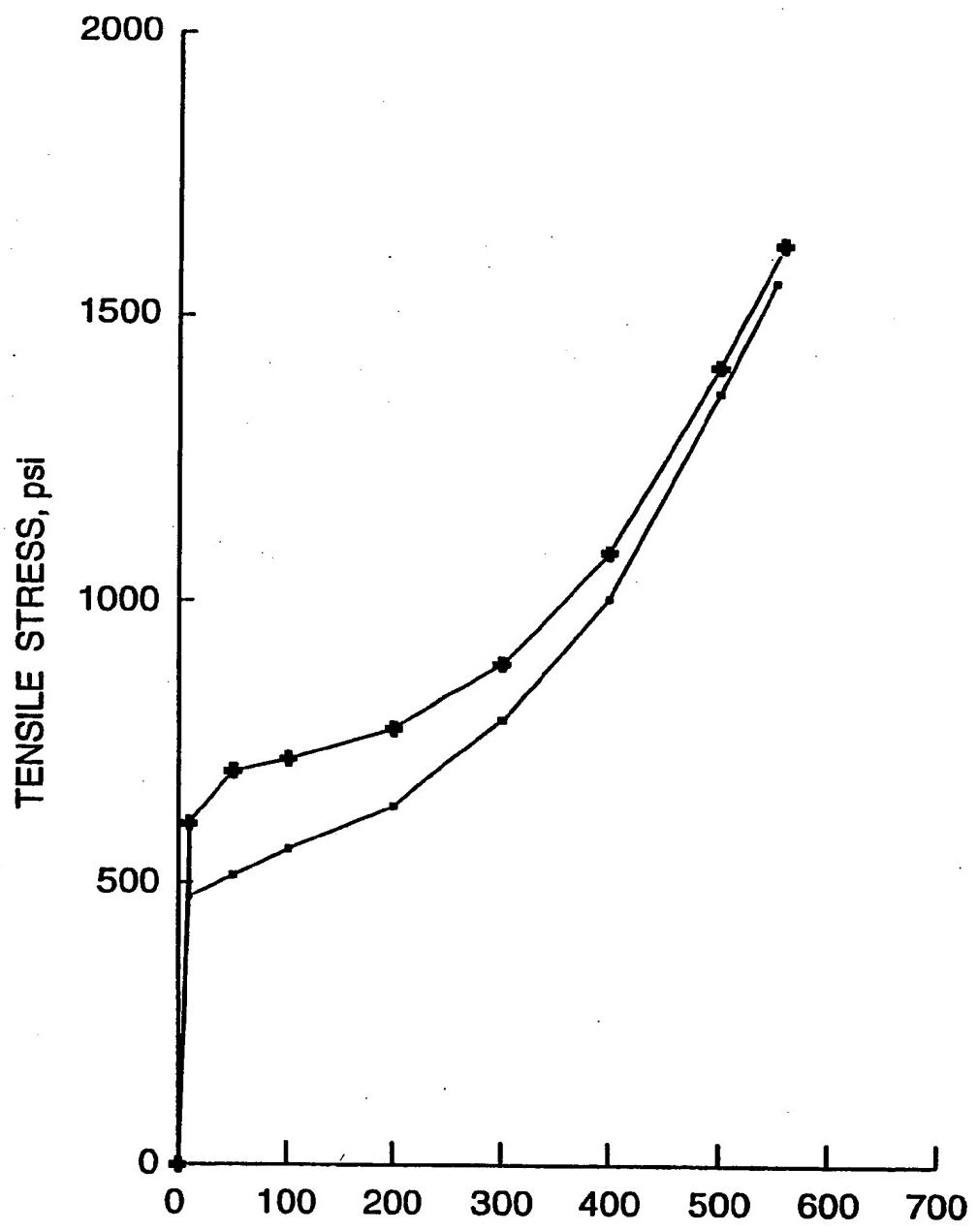


FIG.6d

% ELONGATION

10/18

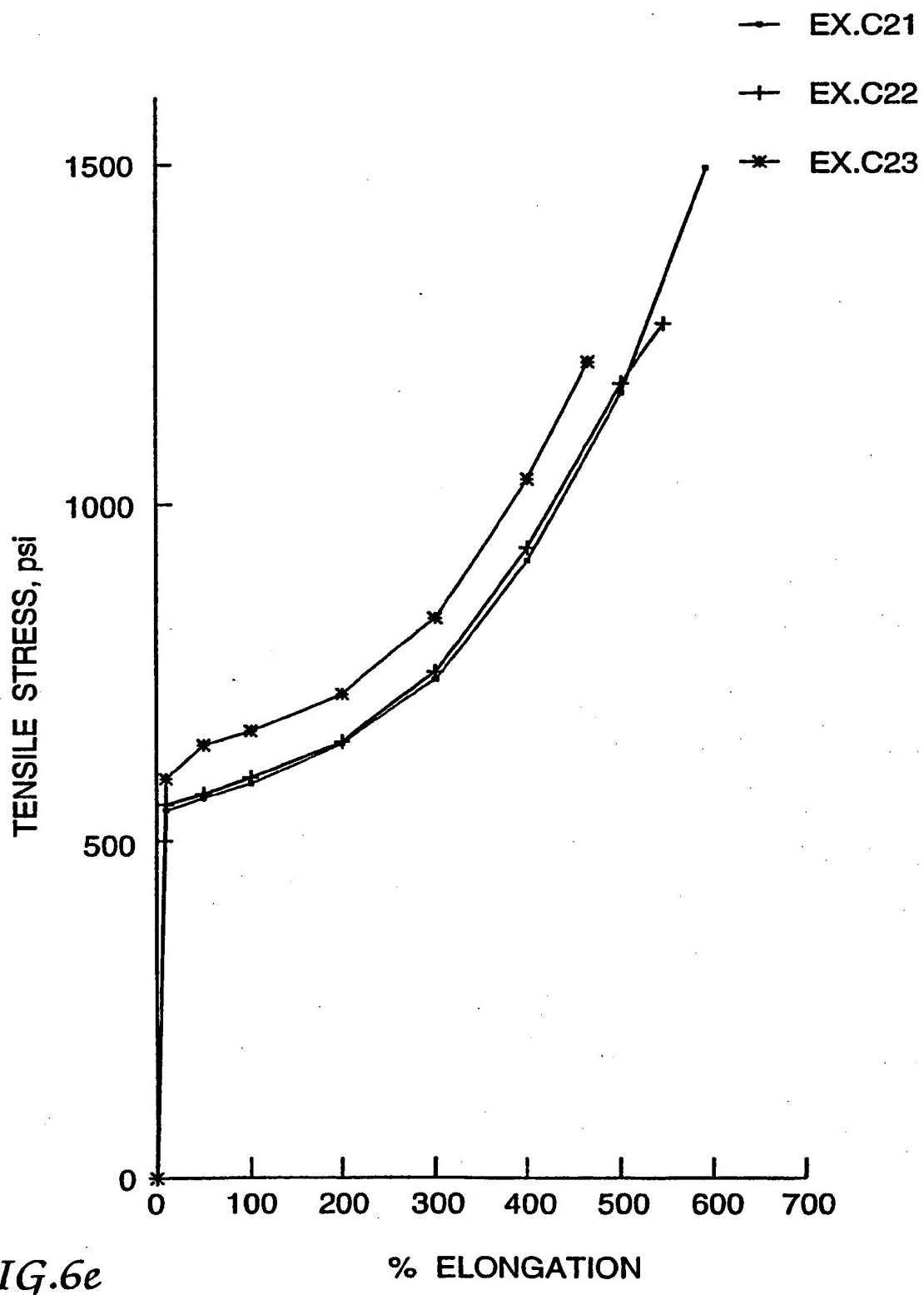


FIG.6e

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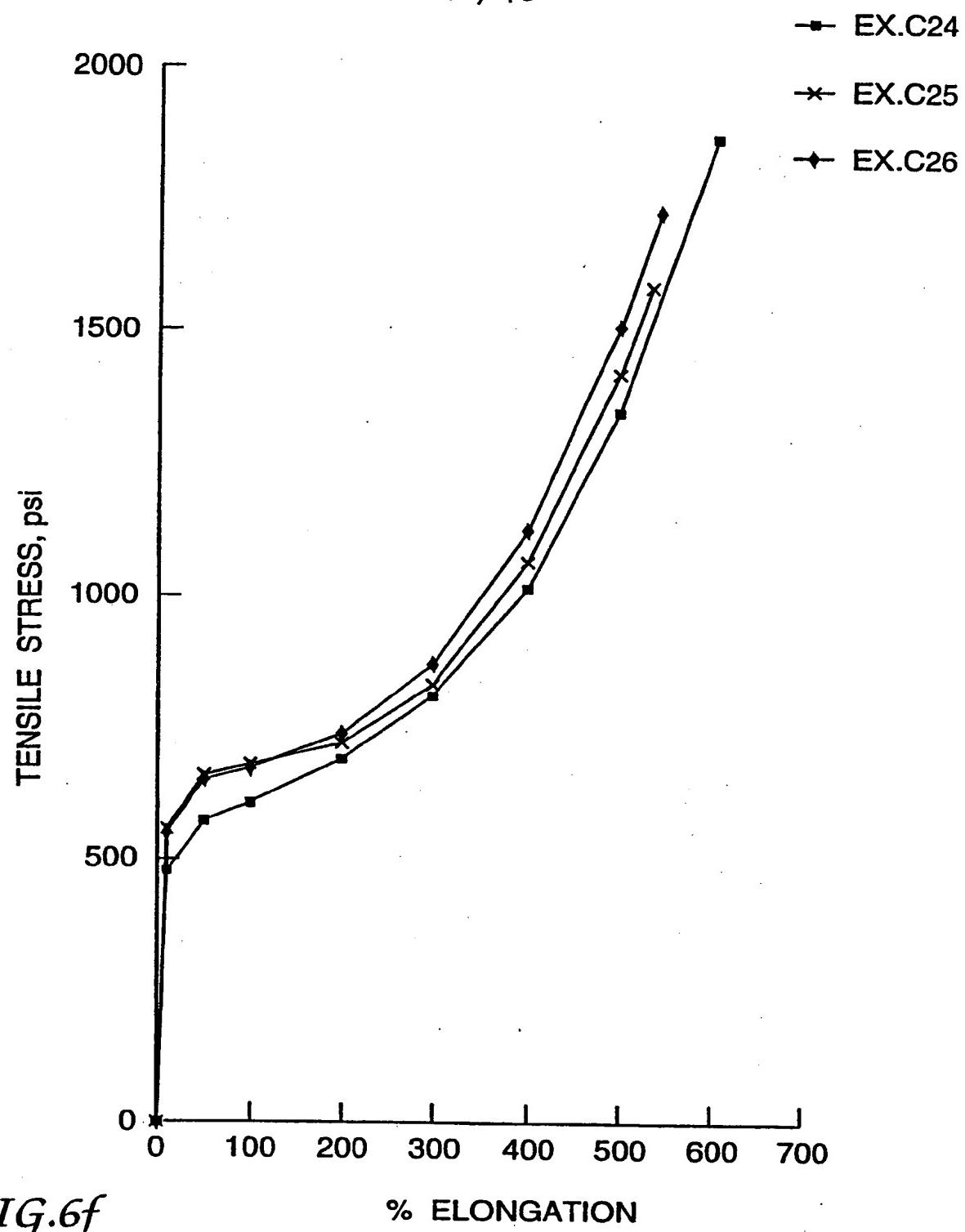


FIG.6f

% ELONGATION

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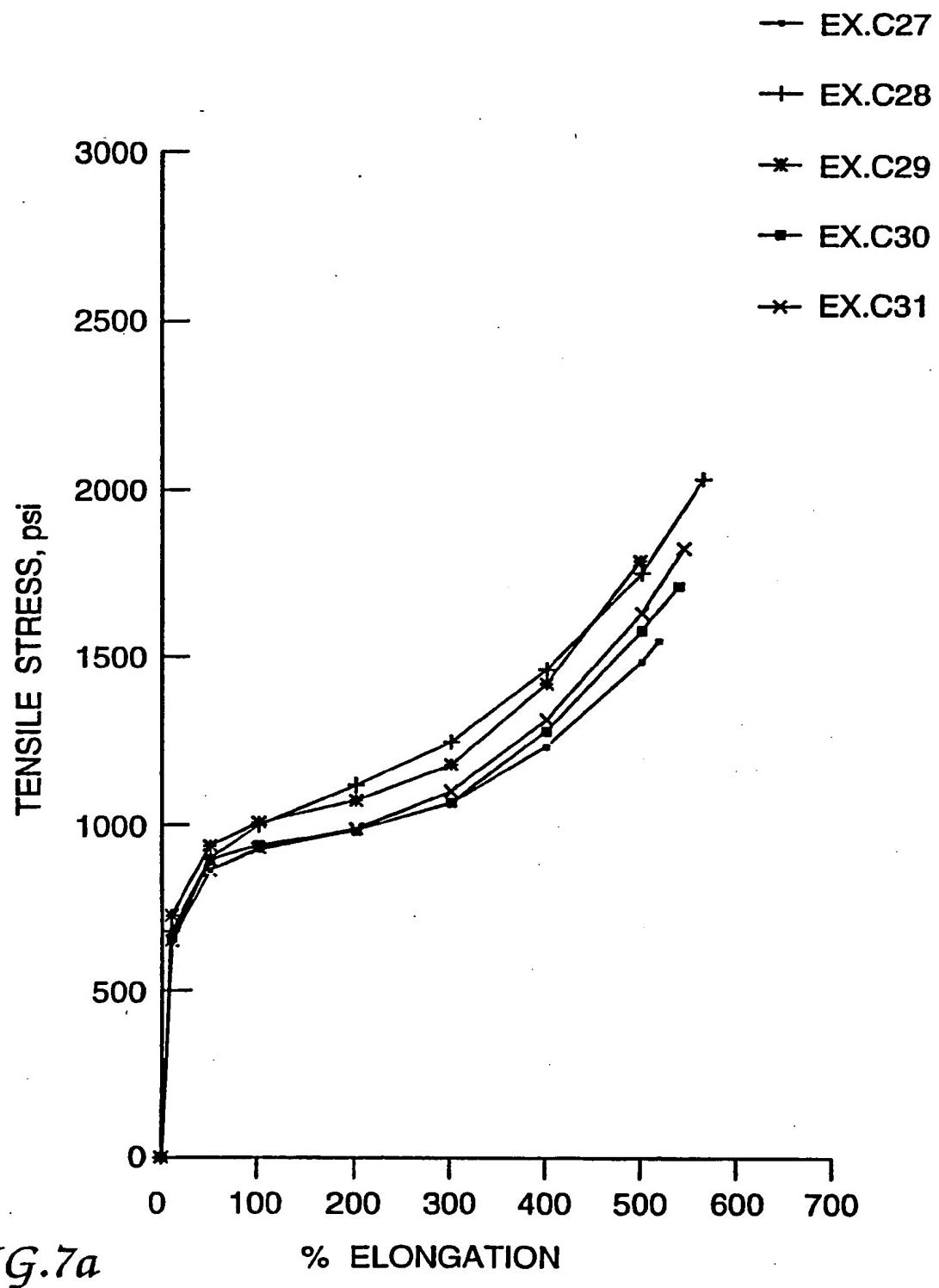


FIG.7a

% ELONGATION

13/18

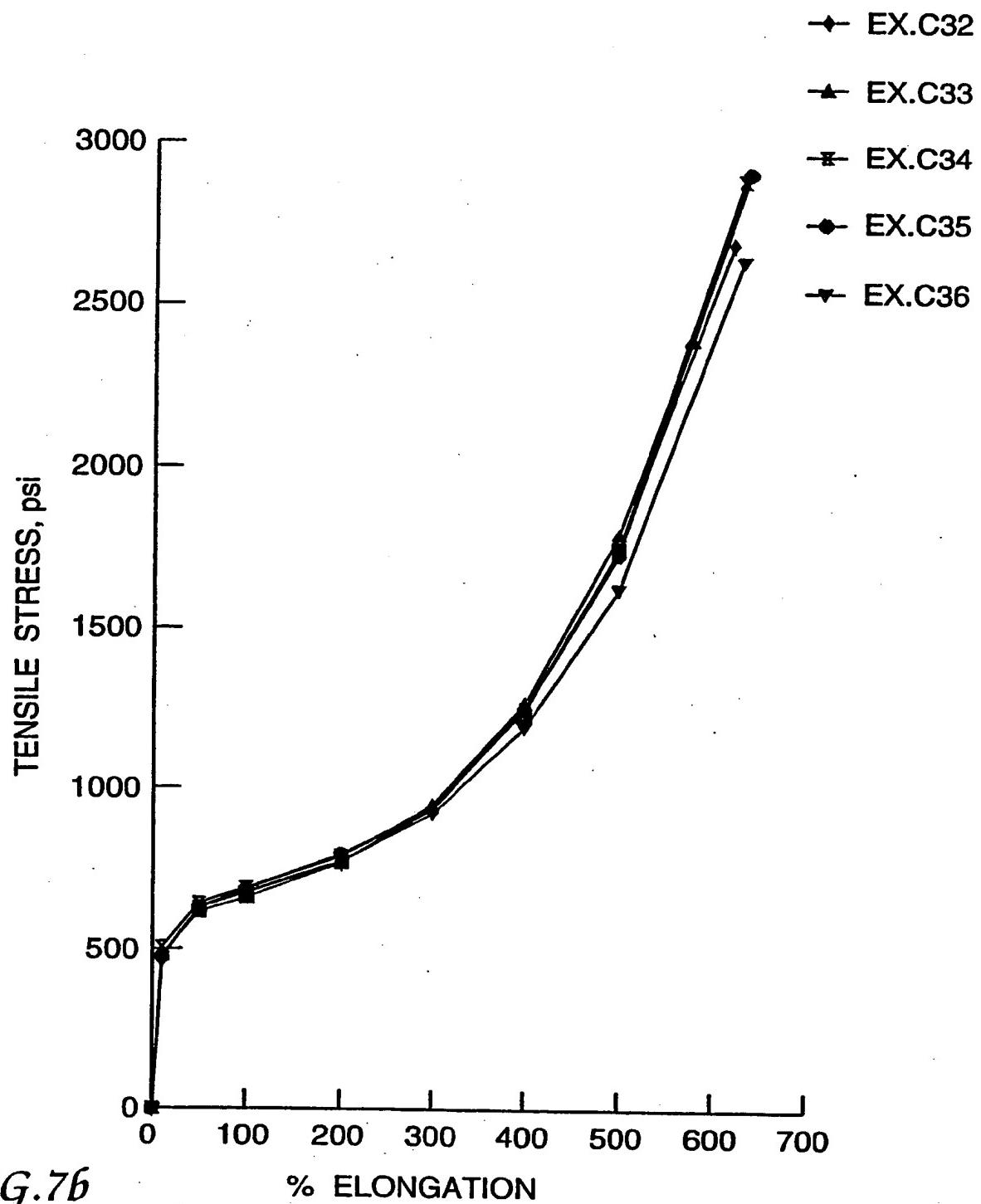


FIG.76

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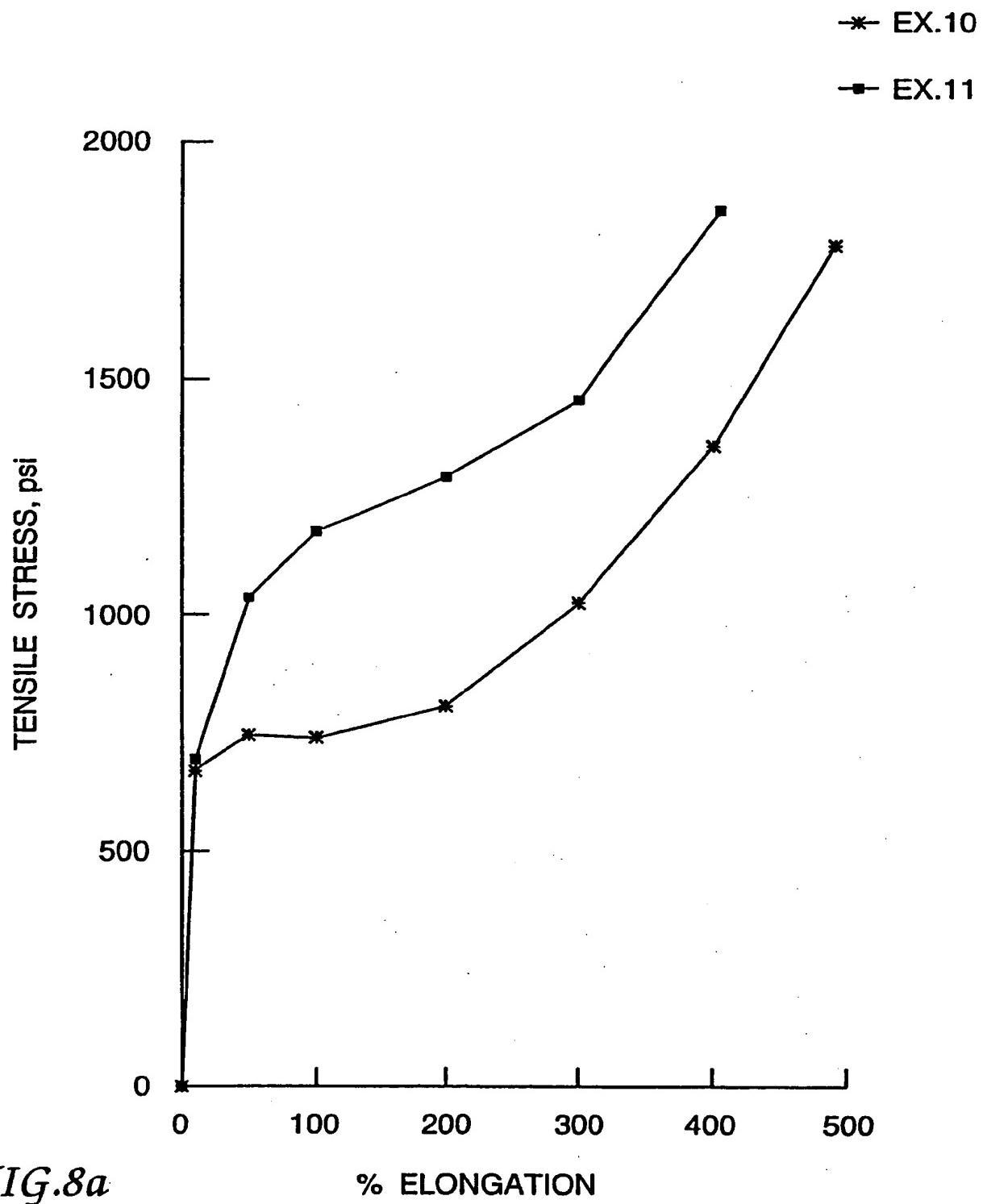


FIG.8a

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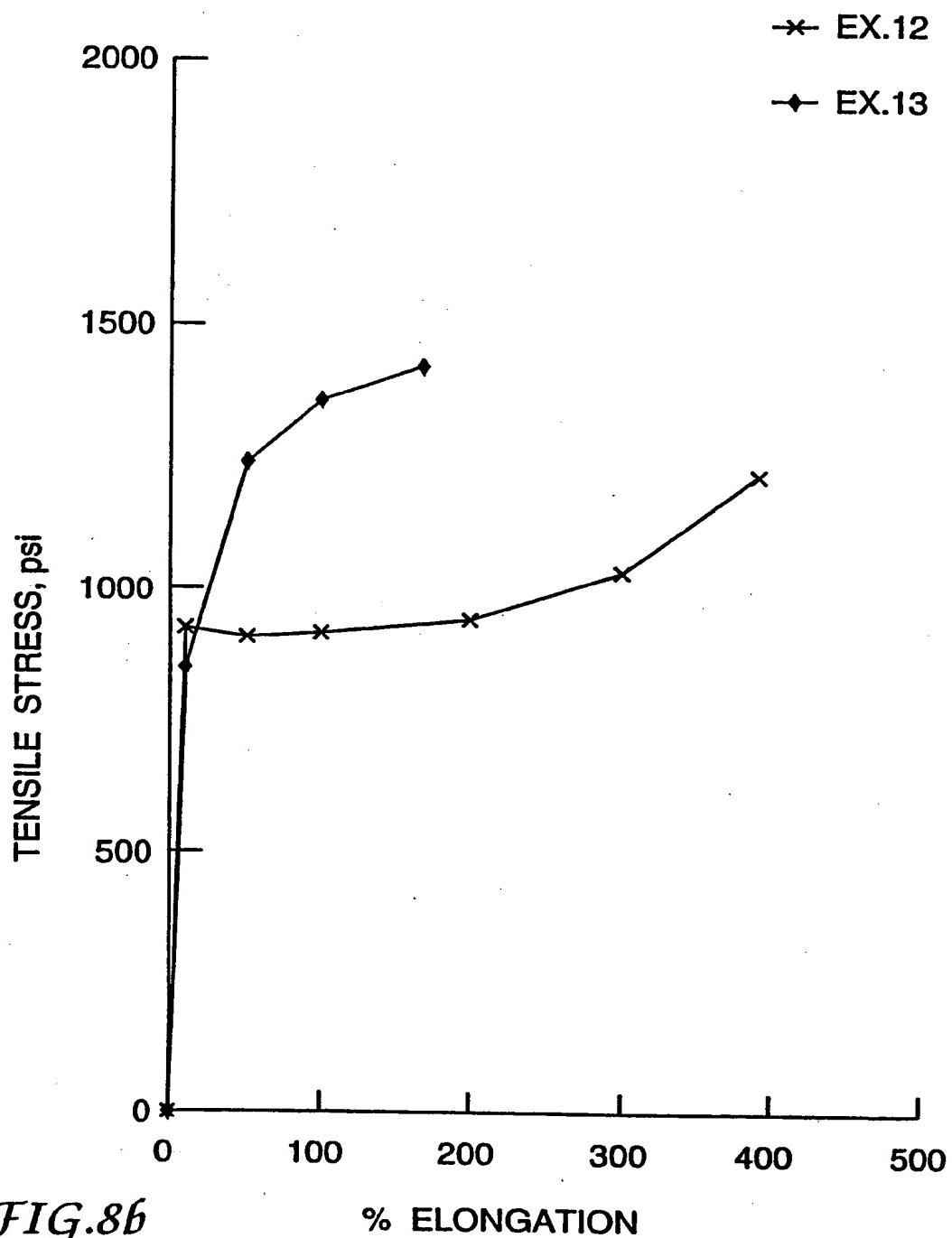


FIG.8b

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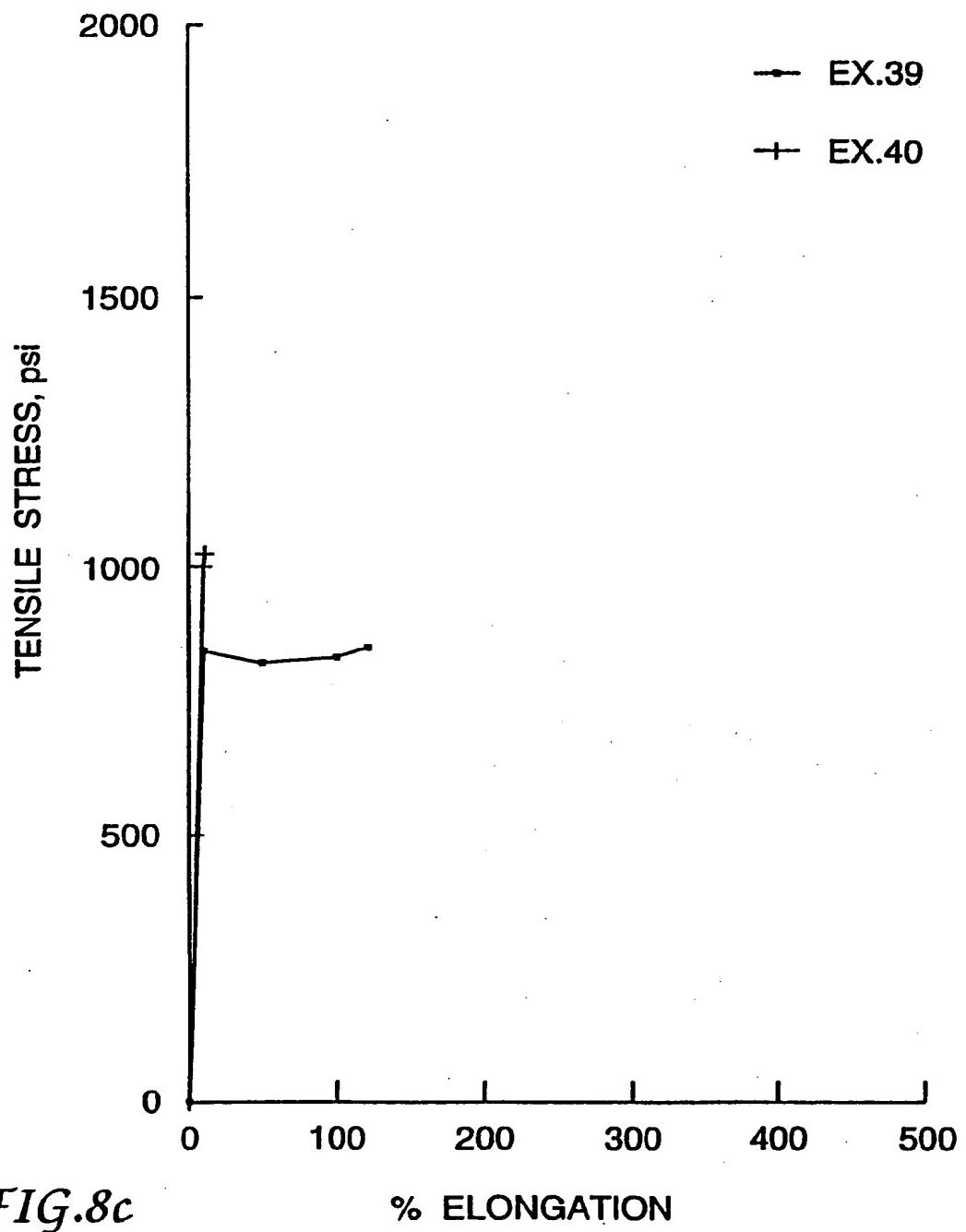


FIG.8c

% ELONGATION

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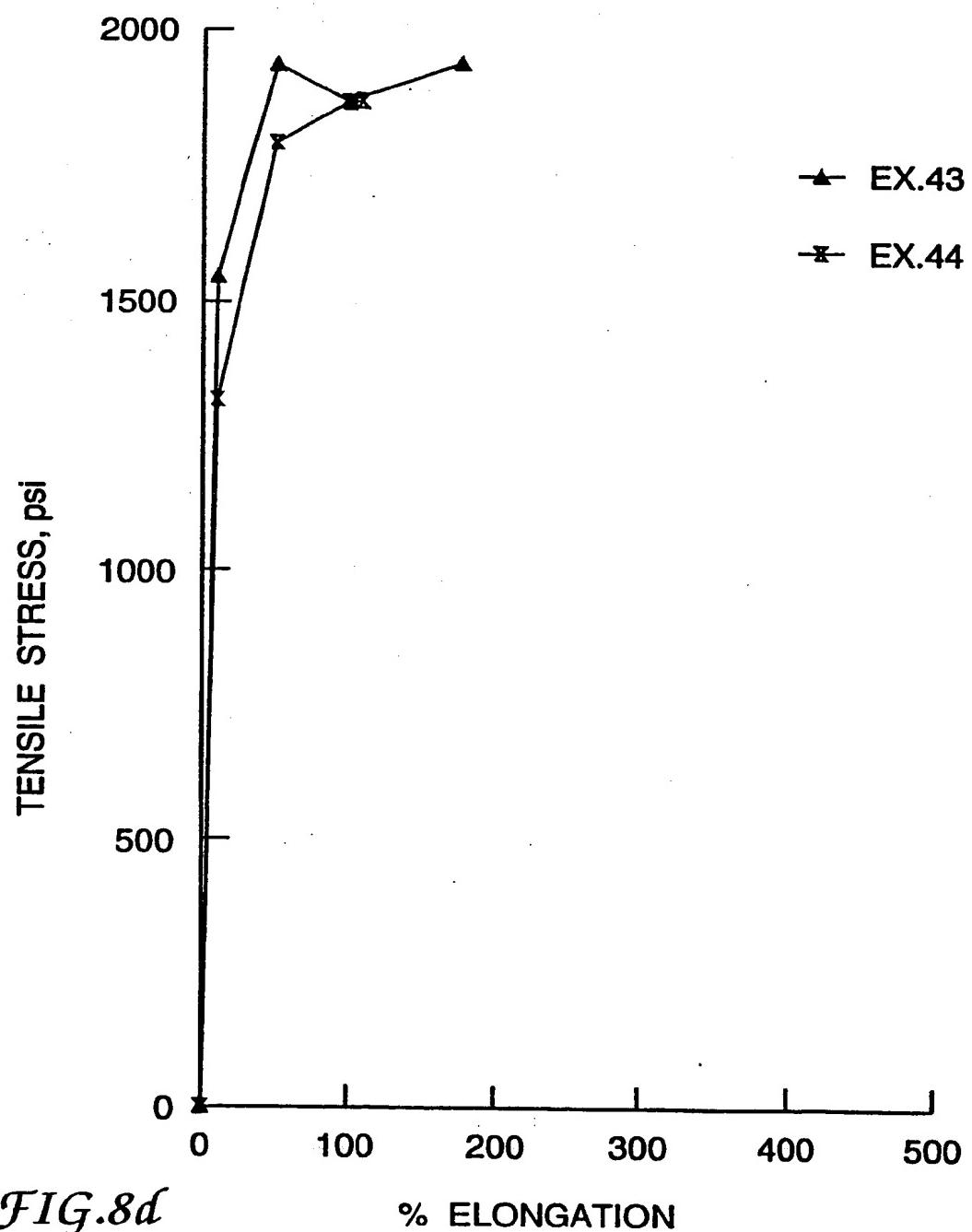


FIG.8d

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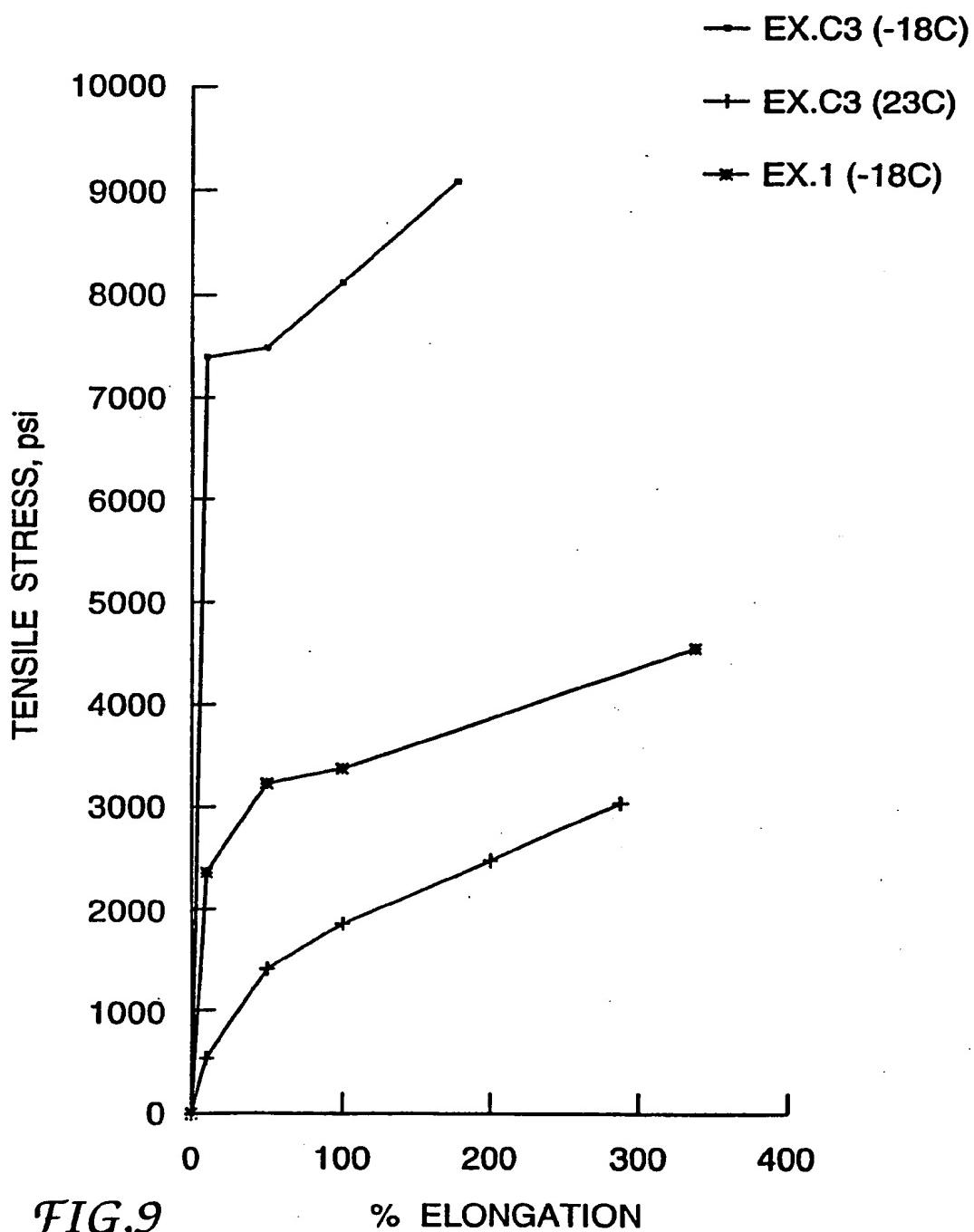


FIG.9

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 94/09171

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 H01B3/44

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 H01B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP,A,0 276 128 (TONEN SEKIYUKAGAKU K.K.) 27 July 1988 ---	
A	US,A,4 798 602 (G.G.LAUS) 17 January 1989 ---	
A	EP,A,0 409 567 (TONEN SEKIYUKAGAKU K.K.) 23 January 1991 ---	
A	EP,A,0 104 350 (BAYER) 4 April 1984 ---	
A	EP,A,0 149 171 (EC ERDOLCHEMIE) 24 July 1985 ---	
A	US,A,5 137 937 (M.T.HUGGARD ET ALL.) 11 September 1992 -----	

<input type="checkbox"/>	Further documents are listed in the continuation of box C.	<input checked="" type="checkbox"/>	Patent family members are listed in annex.
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- *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
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- *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- *Z* document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
29 November 1994	15. 12. 94
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax (+31-70) 340-3016	Authorized officer Stienon, P

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 94/09171

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		WO-A-	9217537	15-10-92

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